New mechanism of first order magnetization process in multisublattice rare-earth compounds

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First-order field-induced spin reorientation transitions in multisublattice intermetallic compounds are considered within an anisotropic Heisenberg model. Unlike previous works, only leading-order anisotropy constants of the sublattices are included. The exchange interactions both between the rare-earth (RE) and transition metal (TM) sublattices and within the RE sublattice play an important role in the model. The latter interaction results in the occurrence of metastable states. One of the minima corresponds to the non-collinear magnetic structure where the RE subsystem is divided into two sublattices, and their moments deviate in opposite directions from the TM moment direction. The possible FOMP scenarios for R_2Fe_{17}, $HoFe_{11}Ti$, and $TbMn_6Sn_6$ systems are discussed.

I. INTRODUCTION

The phenomenon of FOMP (First Order Magnetization Process) was discovered during last years in many intermetallic compounds. Here belong a number of practically important compounds of transition metals with rare-earths (RE) like $R_2Fe_{14}B$ [1,2], $Nd_2Co_{14}B$ [3], R_2Fe_{17} [4–8], $RFe_{11}Ti$ [9–11], RMn_6Sn_6 [12], $Sm_{1-x}Nd_xCo_5$ [13,14], and also some ferrites [15]. In some directions, magnetization jumps with increasing external magnetic field are observed in these systems. Usually this effect is treated phenomenologically as a result of competition of magnetic anisotropy (MA) constants $K^{(n)}$ of different order [16,17]. Presence of large high-order constants enables one to obtain local minima of free energy and thereby first-order transitions. However, such an approach meets with a number of difficulties. Large values of $K^{(n>1)}$ required are not confirmed by crystal-field calculations (see Refs. [1,2,18]). Besides that, high order $K^{(n)}$ are proportional to high powers of magnetization and should rapidly decrease with increasing temperature T. This effect should be stronger in the case where $K^{(n)}$ come mainly from RE sublattice: the rare-earth moments can become rather small at T well below the Curie temperature T_C. The reason is that the exchange interaction $J_{\rm RT}$, which acts on RE ions from the transition metal (TM) system, may be considerably smaller than the interaction $J_{\rm TT}$ which determines T_C [19]. Thus, the local minima explaining FOMP should exist only at low temperatures. However, FOMP is observed as a rule in a wide temperature region [7,12]. The dependences $K^{(n)}(T)$ fitted from magnetization curves turn out to be rather complicated and sometimes seem to be artificial.

The data discussed lead to the conclusion that effects of many-sublattice structure are important in systems demonstrating FOMP [17]. It is known that in a number of intermetallic RE systems (Tm_2Fe_{17}, $RFe_{11}Ti$, $TbMn_6Sn_6$) temperature-induced orientational transitions are caused by competing anisotropy of RE and TM sublattices (in hexaferrites $BaZn_{2-x}Co_xFe_{16}O_{27}$ [15], competition of Fe and Co sublattices plays a similar role). It is natural to suppose that this factor is essential for the field-induced transitions too. Although the many-sublattice case was considered earlier [20], these treatments were also based on the idea of competion of high-order constants.

In the present work we propose a new mechanism of FOMP in a multisublattice magnet, which is connected with the formation of a non-collinear spin structure. The corresponding theoretical model is formulated in Sect.2. In Sect.3 we consider the FOMP picture in various cases and discuss experimental situations.

II. THE THEORETICAL MODEL

We start from the Heisenberg model of uniaxial multisublattice crystal with inclusion of magnetic anisotropy. The MA constants in the problem are usually treated as phenomenological parameters which are obtained by fitting magnetization curves. When describing FOMP one introduces as a rule large high-order constants, $K^{(3)} \sim K^{(2)} \sim K^{(1)}$. To avoid the difficulties discussed above, we include only leading-order MA constants for different magnetic sublattices, $K_i = K_i^{(1)}$. Using the mean-field approximation we write down the expression for the total energy

$$E = \sum_{i>j} J_{ij} \cos(\theta_i - \theta_j) + \sum_i (K_i \sin^2 \theta_i - \mathbf{HM}_i). \quad (1)$$

Here i, j are sublattice indices, $\mathbf{M}_i$ are the corresponding magnetic moments which make the angles θ_i with the z axis, J_{ij} are exchange parameters, $\mathbf{H}$ is the external magnetic field. For the sake of convenience, we have included the dimensionless factors of $M_i = \langle \mathbf{M}_i \rangle$ into J_{ij}, so that $J_{ij} \propto M_i M_j$. The crystal and magnetic structures contain TM and RE sublattices which are coupled by the exchange interaction J_{RT}. The TM subsystem yields usually main contribution to magnetic moment, and the RE one to MA. The most strong interaction J_{TT} plays the role of a mean field in the TM subsystem and needs not to be included explicitly.

As demonstrate crystal-field considerations, formation of non-collinear magnetic structures is possible in multisublattice structures, e.g., for the systems $R_2Fe_{14}B$ [2,1]. The simplest case of a two-sublattice magnet was analyzed earlier in Ref. [19]. It was shown that, in the case where K_{T} and K_{R} have opposite signs (competing anisotropy), the sublattice moments become non-parallel provided that

$$|J_{\mathrm{RT}}(1/K_{\mathrm{R}} + 1/K_{\mathrm{T}})| < 2 \tag{2}$$

If the absolute values of K_{T} and K_{R} are close, this condition can hold even for rather strong exchange J_{RT}.

The two-sublattice problem with non-collinear magnetic structure enables one to obtain a local energy minimum with including magnetic field, but for one field direction only. To avoid this difficulty, we have to introduce, instead of one RE subsystem, *two* equivalent RE sublattices, RI and RII, with $K_{\mathrm{RI}} = K_{\mathrm{RII}} = K_{\mathrm{R}}$, $J_{\mathrm{RIT}} = J_{\mathrm{RIIT}} = J_{\mathrm{RT}}$. This model preserves the correct axial symmetry and yields the energy minimum for both field directions. Of course, for real compounds with complicated crystal structures each 'sublattice' corresponds to several crystal positions. To stabilize the non-collinear structure with split RE sublattice we include the antiferromagnetic exchange interaction between the sublattices, $J_{\mathrm{RR}} > 0$. The J_{RT} and J_{RR} values cannot be reliably obtained from existing experimental data, but for the long-range RKKY exchange interaction $|J_{\mathrm{RR}}|$ may be assumed not small. The essential property of such a model is the presence of metastable states already at $H = 0$. Owing to this fact, first-order phase transitions are possible which occur at crossing the energy levels in the external magnetic field.

III. FOMP SCENARIOS FOR VARIOUS EXPERIMENTAL SITUATIONS

The idea of the FOMP mechanism can be illustrated on a simple toy model where the TM moment is rigidly fixed by strong MA in the x direction (or in the xy plane for the three-dimensional problem). The RE ions which possess more weak easy-axis MA tend to incline to the z direction, each ion having two local minima. The interaction J_{RR} results in that the degeneracy of the minima is lifted, so that the moments of RE sublattices deviate in opposite directions (Fig.1a). With increasing magnetic field along the z axis, a first-order transition occurs to the configuration where RE moments are almost parallel (Fig.1b). A smooth transition does not occur because of the influence of MA of RE ions.

Now we pass to a more realistic situation of finite (and even small) MA of TM subsystem. First we consider the situation where $K_{\mathrm{R}} > 0, K_{\mathrm{T}} < 0$, and the easy-axis MA of RE sublattice predominates. This is the case for the systems Tm_2Fe_{17} and $TbMn_6Sn_6$, as follows from the existence of easy-axis - easy-plane spin-reorientation transitions with increasing temperature ($|K_{\mathrm{R}}|$ decreases more rapidly than $|K_{\mathrm{T}}|$). Two possible metastable states are shown in Fig.2, the exchange J_{RT} being supposed antiferromagnetic, which corresponds to heavy rare-earth ions, e.g., to Tb and Tm. In the case where $J_{\mathrm{RT}} < 0$, which occurs for light rare-earths, e.g., for Nd and Pr, the picture differs by the opposite direction of RE moments.

In the zero magnetic field the first state (Fig.2a) is realized with the collinear spin configuration along the z axis (easy direction for the RE sublattice). At increasing the field $\mathbf{H} \parallel x$, the magnetization vector starts to rotate with retaining the nearly antiparallel orientation of TM and RE moments. For simplicity, we neglect in the analytical treatment the non-collinearity of TM and RE moments for this state, which is weak due to large value of $|J_{\mathrm{RT}}|$. Then the energy in the state (a) is expressed in terms of the angle θ beween the TM moment and z axis,

$$\begin{aligned} E_{\mathrm{a}} &= -2|J_{\mathrm{RT}}| + J_{\mathrm{RR}} - H(M_{\mathrm{T}} \pm 2M_{\mathrm{R}})\sin\theta \\ &\quad + (K_{\mathrm{T}} + 2K_{\mathrm{R}})\sin^2\theta \\ &= -2|J_{\mathrm{RT}}| + J_{\mathrm{RR}} - \frac{(M_{\mathrm{T}} \pm 2M_{\mathrm{R}})^2}{4(2K_{\mathrm{R}} + K_{\mathrm{T}})} H^2 \end{aligned} \tag{3}$$

the sign $+(-)$ corresponding to $J_{\mathrm{RT}} < 0$ ($J_{\mathrm{RT}} > 0$). The second state (Fig.2b) posseses a non-collinear spin configuration with split RE sublattice owing to the gain in the MA energy for the RE subsystem, and also due to the interaction J_{RR}. In this state the spin configuration depends weakly on magnetic field and the TM moment remains parallel to x axis, so that

$$E_{\rm b} = -2|J_{\rm RT}|\cos\alpha + J_{\rm RR}\cos 2\alpha$$
$$-H(M_{\rm T} \pm 2M_{\rm R}\cos\alpha) + K_{\rm T} + 2K_{\rm R}\cos^2\alpha$$
$$= K_{\rm T} - HM_{\rm T} - J_{\rm RR} - \frac{(J_{\rm RT} - HM_{\rm R})^2}{4(J_{\rm RR} + K_{\rm R})} \quad (4)$$

where α ($0 < \alpha < \pi/2$) is the angle of the RE moment deviation from the x axis. The total energy in the state (b) has an almost linear field dependence and decreases more rapidly than that for the state (a) with rotating moment (Fig.3). The angles θ and α are obtained by minimization of Eqs.(3), (4),

$$\sin\theta = \frac{H(M_{\rm T} \pm 2M_{\rm R})}{2(2K_{\rm R} + K_{\rm T})}, \cos\alpha = \frac{|J_{\rm RT} - HM_{\rm R}|}{2(J_{\rm RR} + K_{\rm R})}, \quad (5)$$

Thus the local minimum (non-collinear state) occurs at

$$J_{\rm RR} + K_{\rm R} > |J_{\rm RT} - HM_{\rm R}|/2. \quad (6)$$

Note that the minimum can occur with including magnetic field, even if it is a absent at $H = 0$. The transition field and the angle α (but not the magnetization jump) turn out to be small provided that $2|J_{\rm RR} + K_{\rm R}| \simeq |J_{\rm RT}|$. Since $J_{\rm RR}$ and $K_{\rm R}$ are proportional to $M^2(T)$ (unlike the constants $K^{(n>1)}$ in standard FOMP considerations, which decrease rapidly with increasing T), the criterion (6) depends rather weakly on T. Therefore the mechanism discussed enables one to obtain FOMP in a wide temperature region.

The numerical calculations were performed by direct minimization of total energy (1) without approximations discussed above. The moment values were taken to be $M_{\rm T} = 4, M_{\rm RI} = M_{\rm RII} = M_{\rm R}/2 = 1$, which corresponds roughly to R_2Fe_{17} compounds (e.g., for Tb_2Fe_{17} the moment of Fe subsystem exceeds by two times that of RE subsystem, $M_{\rm Tb} = 18.6\mu_B, M_{\rm Fe} = 36.6\mu_B$ per formula unit [5]). At some critical field $E_{\rm a}$ becomes equal to $E_{\rm b}$ (Fig.3), and the magnetization demonstrates a vertical jump (Fig.4). It should be noted that the transition field is much smaller than the exchange interaction $J_{\rm RR}$, unlike the situation of Fig.1. After the jump, the magnetization increases rather slowly because of large value of $J_{\rm RT}$. On the other hand, the magnetization depends very weakly on the field $\mathbf{H} \parallel z$ because of the strong interaction $J_{\rm RT}$. Owing to the deviation from the antiparallel alignment, the magnetization in the hard direction x after the transition turns out to be larger than the zero-field magnetization in the easy direction z ($M_x > M_z = 4 - 2 = 2$). Such a behavior is demonstrated by experimental dependences $M(H)$ in a number of systems, e.g., in Tm_2Fe_{17} [7,8].

The zero-field canting angle α of the RE sublattice for the parameters of Fig.3 makes up about 20^0. Such small values do not contradict likely experimental data. Indications of formation of the non-collinear structure after the transition are given by neutron scattering data on Tm_2Fe_{17} [8] and $TbMn_6Sn_6$ [21]. Non-collinear structures with the split RE sublattice were also observed for $R_2Fe_{14}B$ systems [22].

In the system Tb_2Fe_{17} [5,7] the Fe sublattice has the easy-plane magnetic anisotropy, as well as the Tb sublattice (although Tb ions in different positions are supposed to have MA of different signs). Thus, the competing anisotropy situation does not occur. Nevertheless, in this case our approach also enables one to obtain FOMP, but the transition field turns out to be somewhat larger. Fig.5 shows the result of calculation of magnetization for the constants of the same sign, $K_{\rm R} > 0, K_{\rm T} > 0$ (the easy-plane and easy-axis problems are equivalent provided that we restirict ourselves to the plane model). One can see that before the jump the field dependence of the magnetization is practically linear, unlike Fig.4. The state with split RE sublattices occurs only with increasing magnetic field.

The curves obtained demonstrate a similarity to experimental ones. Let us try to perform a more detailed comparison with experimental data. The critical FOMP field is determined by the overall scale of the anisotropy parameters. Unfortunately, MA constants are usually fitted from magnetization curves with including high-order constants which contradicts our model. Therefore we shall concentrate on the cases where the parameters of the model can be determined from independent experiments. Using the data for Tm_2Fe_{17} from Ref. [8], $M_{\rm Tm} \simeq 12\mu_B, M_{\rm Fe} \simeq 30\mu_B$ per formula unit, $J_{\rm TmFe} = 60{\rm J/cm}^3$, $K_{\rm Fe} = -2.5$ J/cm^3, $K_{\rm Tm} = 7$ J/cm^3, we can fit the experimental FOMP field, $\mu_0 H = 5$T, by taking $J_{\rm RR}/J_{\rm RT} \simeq 0.5$. Unfortunately, the value of the magnetization jump cannot be reliably determined from the results of [7,8].

The values of MA constants for Tb_2Fe_{17} can be estimated from NMR data [23] (see Ref. [24]), $K_{\rm Tb} = -10$ J/cm^3, $K_{\rm Tb} = 3$ J/cm^3 for the positions 2b and 2d, $K_{\rm Fe} = -3$ J/cm^3. Of course, the accuracy of these values is rather poor. After averaging the values of $K_{\rm Tb}$ over sublattices such a relation between MA constants corresponds roughly to the parameters of Fig.5. The rescaled FOMP critical field from this Figure turns out to be by about two times larger than the experimental value which makes up $\mu_0 H \simeq 4$T. The agreement can be made better provided that we take somewhat smaller values of $|K_{\rm Fe}|$ and increase the ratio $J_{\rm RR}/J_{\rm RT}$. Besides that, the situation in Tb_2Fe_{17} is complicated because of existence of two FOMP-type transitions [7].

Now we treat the case where easy-axis anisotropy $K_{\rm T}$ dominates over MA of RE sublattice, which has an opposite sign (such a stituation takes place, e.g., for the compound $HoFe_{11}Ti$). Then the qualitative picture of FOMP turns out to be different from Fig.2. As shown in Fig.6a, at $H = 0$ RE moments deviate from the axis z (easy direction for TM sublattice) by the angle

$$\alpha = \arccos\left[\frac{|J_{\rm RT}|}{2(J_{\rm RR} - K_{\rm R})}\right]. \tag{7}$$

This configuration begins to rotate when including the small magnetic field in the x direction. At further increasing the field, the nearly collinear spin configuration shown in Fig.6b becomes favorable. As follows from (7), FOMP is possible at $J_{\rm RR} - K_{\rm R} > |J_{\rm RT}|/2$. Note that the solution (local minimum), corresponding to the non-collinear structure, can vanish with increasing H somewhat above the critical field of FOMP. A structure of the type Fig.6a (but with ferromagnetic interaction $J_{\rm RT} < 0$) was proposed for $Y_{1-x}Nd_xCo_5$ on the basis of magnetic measurements [25].

After the jump, the magnetization in the x direction continues to increase rather rapidly due to rotation of TM moments (Fig.6b), and becomes saturated only when the moments lie along the x axis ($M_x = 2$). Absence of the saturation immediately after the jump corresponds to FOMP-II according to the classification [16,17]. The result of magnetization calculation is shown in Fig.7. For simplicity, we restrict ourselves again to the plane problem (xz plane model). The parameter values are chosen to demonstrate existence of the saturation region, although FOMP can be obtained for smaller $|K_i/J_{\rm RT}|$ and at smaller H too. The dependence obtained is in a qualitative agreement with that for $HoFe_{11}Ti$ [11]. This system is close to instability with respect to the formation of the 'easy-cone' structure [11] owing to competing MA of Ho and Fe sublattices with $K_{\rm R} < 0, K_{\rm T} > 0$.

IV. CONCLUSIONS

To conclude, we have proposed a simple model which reproduces qualitatively the FOMP pictures in some RE systems with different sign combinations of sublattice MA constants. It should be noted that the field-induced transitions considered are similar in some respects to the spin-flop transitions in anisotropic antiferromagnets. In the latter situation, formation of metastable states and magnetization jumps are possible too (see, e.g., Ref. [26]). However, our picture is more complicated since this includes the TM sublattice.

The consideration performed enables one to avoid some difficulties and reduce the number of fitting parameters in comparison with the treatments that use high-order MA constants. Further experimental investigations of magnetic structures, especially with the use of neutron scattering, would be of interest to verify the FOMP scenarios proposed.

When considering the situation in other compounds, we meet with some problems. The description of FOMP for the systems $Pr_2Fe_{14}B$, $Nd_2Fe_{14}B$ should also include non-collinear structures [2], but this problem is more difficult. In the compound $Nd_2Fe_{14}B$ a conical structure occurs in zero field, although the signs of MA for Nd and Fe sublattices coincide. Usually such a behavior is attributed to the influence of higher-order anisotropy constants; possibility of different role of various Nd positions is also discussed. It should be noted that the FOMP picture is similar for the compound $Nd_2Co_{14}B$ [3] where the Co ions have easy-plane MA. For the system $Sm_{1-x}Nd_xCo_5$ [13] the RE sublattice already includes two subsystems with opposite MA signs. However, to obtain FOMP in spirit of our mechanism one still has to divide this sublattice into two symmetric parts.

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FIGURE CAPTIONS

Fig.1. Schematic picture of FOMP in a simple toy model with ferromagnetic exchange $J_{\rm RT}$, $K_{\rm R} > 0$ and large easy-plane $K_{\rm T} < 0$ (a) the spin configuration in the zero magnetic field (b) the spin configuration after the transition induced by the magnetic field in the x direction.

Fig.2. Schematic picture of FOMP in the case of antiferromagnetic exchange $J_{\rm RT}, K_{\rm R} > 0$ (a) the spin configuration in the zero field (b) the non-collinear spin configuration after the magnetization jump induced by the magnetic field $\mathbf{H} \parallel x$.

Fig.3. The field dependence of the total energy in units of $J_{\rm RT} = 1$. Other parameter values are $K_{\rm R} = 0.1, K_{\rm T} = -0.15, J_{\rm RR} = 0.5$. Long-dashed line corresponds to the state (a) in Fig.2, and the short-dashed line to the state (b).

Fig.4. The field dependence of the magnetization in the x direction for the same parameters as in Fig.3. The observable dependence demonstrating FOMP (vertical jump) is shown by the solid line.

Fig.5. The magnetization curve in the x direction for $J_{\rm RT} = 1, K_{\rm R} = 0.1, K_{\rm T} = 0.1, J_{\rm RR} = 0.6$.

Fig.6. Schematic picture of FOMP in the case of antiferromagnetic exchange $J_{\rm RT}$, $K_{\rm R} < 0$. The spin configuration is non-collinear before the jump (a) and becomes collinear after the jump (b).

Fig.7. The magnetization curve in the x direction for $J_{\rm RT} = 1, K_{\rm R} = -0.2, K_{\rm T} = 0.4, J_{\rm RR} = 0.45$. Long-dashed line corresponds to the state (a) in Fig.6, and the short-dashed line to the state (b).

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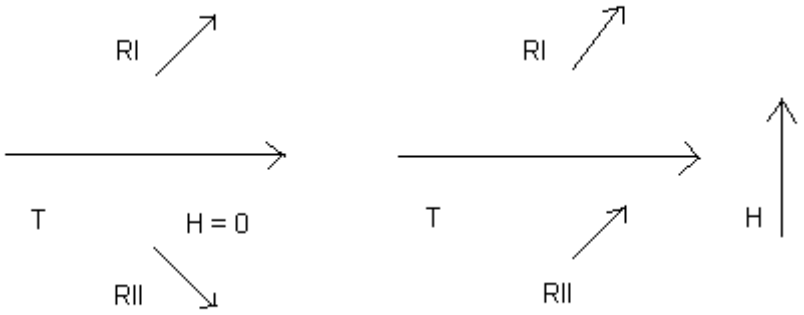
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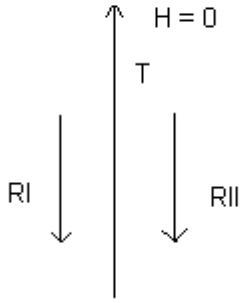

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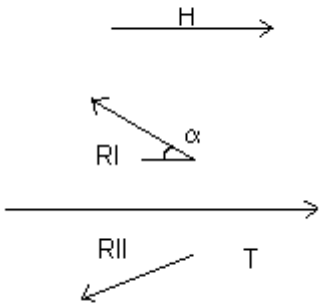

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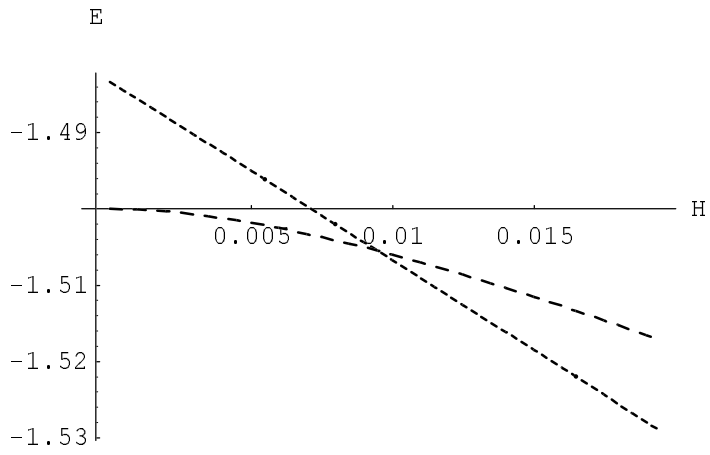
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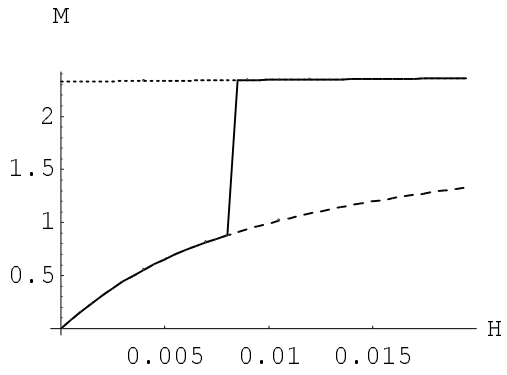
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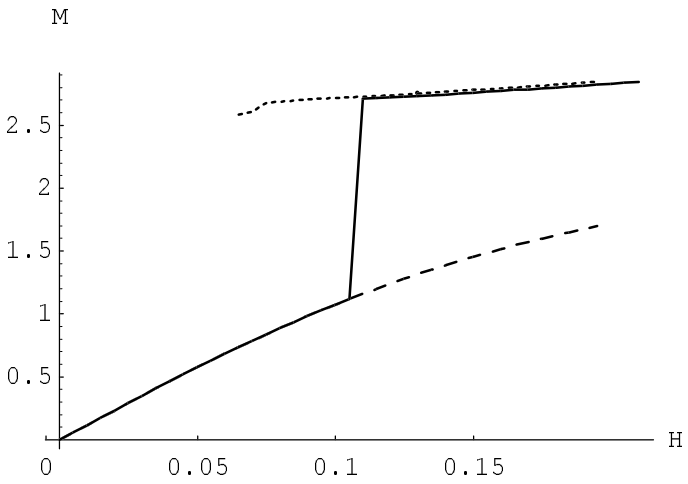
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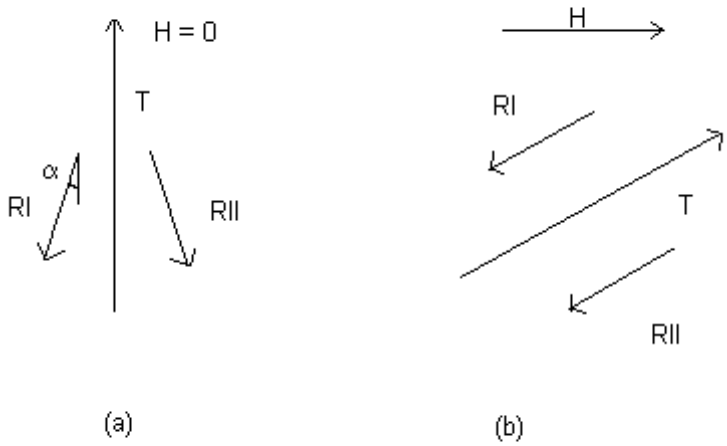

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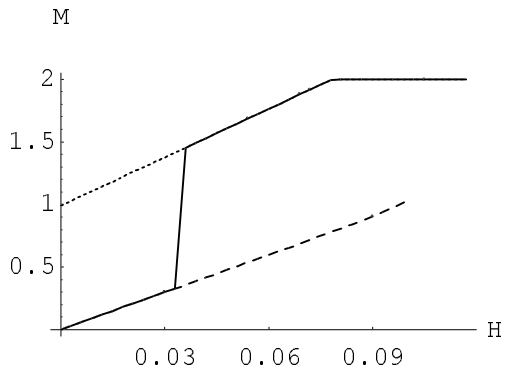
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